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# How shear flow of a semidilute suspension modifies its self-mobility

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We investigate a semidilute hard sphere colloidal suspension with no hydrodynamic interactions. The suspension undergoes a steady shear flow. In addition, a small constant external force induces a tagged particle flux. In order to determine the shear-dependent self-mobility matrix (relating the flux to the force) we construct the approximate solution to the two-particle Smoluchowski equation at contact, applying the induced source method, combined with a power expansion in the square root of the shear rate. We evaluate the self-mobility tensor numerically by means of Padé approximants for small and moderate shear rates.

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## I. INTRODUCTION

Colloidal suspensions under steady shear undergo pronounced structural changes revealed by a distortion of the structure factor from its equilibrium form [1–5]. The magnitude of this distortion is determined by the competition between Brownian motion, driving system towards equilibrium, and the particle convective motion due to shear. The structural changes are accompanied by changes of the suspension transport properties [1, 6]. In particular, for low densities viscosity often decreases with increasing shear rate [7–12]; this phenomenon is known as shear thinning and stems from changes of the two-particle distributions induced by the shear flow. At higher particle volume fractions the opposite relation has been observed, i.e., the increase of viscosity with increasing shear [13, 14, 12]. This phenomenon, known as shear thickening, has been attributed to the formation of large particle aggregates. Particle mobility and diffusion [15–17] are also influenced by the shear flow. In particular, the Einstein relation between effective self-diffusion and self-mobility coefficients does not hold for a nonzero shear rate [18]. A full theoretical understanding of the above-mentioned phenomena is still lacking.

Recently, Bławzdziewicz and Szamel [11–20] proposed a method to evaluate the structure factor and the viscosity of a sheared semidilute hard-sphere suspension with no hydrodynamic interactions, in a range of small and moderate shear rates. In this paper we outline how to modify their procedure to calculate the steady-state shear-rate-dependent self-mobility for the same suspension model. A detailed presentation of our results will be given elsewhere [21, 22]. Self-diffusion process can also be treated by a similar method.

## II. SYSTEM AND EQUATIONS

The suspension consists of mechanically identical colloidal particles of radius $a$, interacting via the hard-sphere potential, with no hydrodynamic interactions. Some of the particles are tagged; their density $n_s$ is uniform and much smaller than the uniform density of untagged particles $n$. A uniform external force $\vec{F}_{\rm ext}$ is applied to tagged particles.

Let $\vec{r}_1$ and $\vec{r}_2$ denote the position of the tagged and an untagged particle, respectively; $\vec{r} = \vec{r}_1 - \vec{r}_2$ is their relative position. The suspension is semidilute, i.e., the volume fraction of untagged particles $\phi = \frac{4}{3}\pi a^3 n$ is sufficiently low for neglecting three-particle contributions to the two-particle density: $n_2(\vec{r}_1, \vec{r}_2, t) \approx n_s n \, g_2(\vec{r}, t)$, where $g_2$ is the low density two-particle distribution, independent of $n$.

The suspension undergoes a steady shear flow

$$\vec{v}(\vec{r}) = \gamma_0 y \hat{e}_x, \tag{1}$$

with a constant shear rate $\gamma_0$. Suspended particles perform Brownian motion. The system is described by the two-particle stationary Smoluchowski equation for two-particle distribution $g_2(\vec{r})$ [1, 6]

$$\vec{\nabla} \cdot \vec{j}_2(\vec{r}) = 0, \qquad r > 2a \tag{2}$$

with a boundary condition on $g_2(\vec{r})$ at contact

$$\vec{r} \cdot \vec{j}_2(\vec{r}) \Big|_{r=2a} = 0 \tag{3}$$

and the two-particle current density $\vec{j}_2(\vec{r})$ taken as

$$\vec{j}_2(\vec{r}) = (-2D_0 \vec{\nabla} + \mu_0 \vec{F}_{\rm ext} + \gamma_0 y \hat{e}_x) \, g_2(\vec{r}) \, . \tag{4}$$

In addition, $g_2(\vec{r}) \to 1$ for $r \to \infty$. The diffusion constant $D_0$ is related to the mobility of an isolated particle $\mu_0$ through the Einstein relation $D_0 = k_B T \mu_0$.

The external force $\vec{F}_{\rm ext}$ generates $\vec{j}_s$, the tagged particle flux relative to the shear flow, in general not parallel to $\vec{F}_{\rm ext}$:

$$\vec{j}_s = n_s \overleftrightarrow{\mu} \, \vec{F}_{\rm ext}. \tag{5}$$

Equation (5) defines the generalized self-mobility tensor $\overleftrightarrow{\mu}$. According to the low density assumption, $\overleftrightarrow{\mu}$ is a linear function of $\phi$,

 

$$\overleftrightarrow{\mu} = \mu_0 \left( \overleftrightarrow{I} + 2\phi \overleftrightarrow{M} \right), \tag{6}$$

where $\overleftrightarrow{I}$ is the unit tensor and $\overleftrightarrow{M}$ is the two-particle contribution to self-mobility. Our goal is to determine the dependence of $\overleftrightarrow{M}$ on the shear rate $\gamma_0$. In general, $\overleftrightarrow{M}$ would also depend on the external force; however, in Sec. IV we take $\vec{F}_{\text{ext}}$ sufficiently small to allow for the linearization of the boundary problem (2)–(4). Within this approximation $\overleftrightarrow{M}$ is independent of $\vec{F}_{\text{ext}}$.

For hard spheres with no hydrodynamic interactions $\vec{j}_s$ is determined by $g_2$ as follows:

$$\vec{j}_s = n_s \mu_0 \left( 4a^2 n k_B T \int_{S^2} d^2\Omega\, \hat{r}\, g_2(2a\hat{r}) + \vec{F}_{\text{ext}} \right), \tag{7}$$

with $\hat{r} = \vec{r}/r$. The dynamics of the system is characterized by the following dimensionless parameters (Péclet numbers):

$$\vec{F} = \frac{a}{2k_B T} \vec{F}_{\text{ext}}, \tag{8}$$

$$\gamma = \frac{a^2}{2D_0} \gamma_0. \tag{9}$$

$\vec{F}$ and $\gamma$ are equal to the ratio of the corresponding characteristic time scales. Namely, $\gamma = \tau_b/\tau_s$ and $F^i = \tau_b/\tau_d^i$, $i = x, y, z$, where $\tau_b = a^2/2D_0$, $\tau_s = 1/\gamma_0$, and $\tau_d^i = a/\mu_0 F_{\text{ext}}^i$ are the time scales describing Brownian motion, shear flow, and force drift, respectively. We assume that $F^i \ll 1$ and we linearize the problem in $\vec{F}$. To evaluate self-mobility for small and moderate $\gamma$ we use the series expansion in $\gamma^{1/2}$; our numerical results are accurate for $\gamma \leq 5$.

It is convenient to introduce dimensionless space coordinates $\vec{R} \equiv \vec{r}/2a$ and to use the corresponding dimensionless two-particle distribution $G_2(\vec{R}) \equiv g_2(\vec{r})$ and dimensionless fluxes $\vec{J}_s(\vec{R}_1) \equiv \frac{a}{2D_0} \vec{j}_s(\vec{r}_1)$ and $\vec{J}_2(\vec{R}) \equiv \frac{a}{2D_0} \vec{j}_2(\vec{r})$. To calculate the self-mobility tensor $\overleftrightarrow{\mu}$ from Eqs. (5) and (7) it is sufficient to evaluate $G_2$ at contact, i.e., on the unit sphere only. A method to evaluate $G_2(\hat{R})$, the solution to the boundary problem (2)–(4) at the unit sphere $\vec{R} = \hat{R}$, is developed in Secs. III–V.

## III. INDUCED SOURCE METHOD

The idea is to construct the solution $G_2(\vec{R})$ to the boundary problem (2)–(4) (rewritten in dimensionless variables) as a linear combination of fundamental solutions $T_{\alpha\beta\zeta}(\vec{R})$ to Eq. (2). $T_{\alpha\beta\zeta}$ correspond to the multipole sources $(-1)^{\alpha+\beta+\zeta} \frac{\partial^\alpha}{\partial X^\alpha} \frac{\partial^\beta}{\partial Y^\beta} \frac{\partial^\zeta}{\partial Z^\zeta} \delta^3(\vec{R})$, with $\alpha, \beta$ non-negative integers and $\zeta = 0, 1$ only (for $\zeta > 1$, $T_{\alpha\beta\zeta}$ are linearly dependent on $T_{\kappa\eta\rho}$ with $\rho = 0, 1$ [22]). The coefficients are determined by the boundary condition (3).

Similarly as in [11, 19], the fundamental solutions $T_{\alpha\beta\zeta}$ can be evaluated [22] from the relation

$$T_{\alpha\beta\zeta}(\vec{R}) = \int_0^\infty d\tau \left( -\frac{\partial}{\partial X} \right)^\alpha \left( -\frac{\partial}{\partial Y} - 2\tau \frac{\partial}{\partial X} \right)^\beta \times \left( -\frac{\partial}{\partial Z} \right)^\zeta P(\vec{R}, \tau), \tag{10}$$

where $P$ [23] is the solution to the time-dependent Smoluchowski equation with $\gamma^{-1}\delta^3(\vec{R})$ as the initial condition ($\tau = t\gamma_0/2$ is rescaled time),

$$P(\vec{R}, \tau) = \frac{\gamma^{1/2}}{\left(1 + \frac{\tau^2}{3}\right)^{1/2} (2\pi\tau)^{3/2}} \times \exp\left\{ -\frac{\gamma}{2\tau} \left[ \frac{\left( X - F_x \frac{\tau}{\gamma} - \tau Y \right)^2}{1 + \frac{\tau^2}{3}} + \left( Y - F_y \frac{\tau}{\gamma} \right)^2 + \left( Z - F_z \frac{\tau}{\gamma} \right)^2 \right] \right\}. \tag{11}$$

We express the solution to the boundary problem (2)–(4) as a linear combination of fundamental solutions

$$G_2(\vec{R}) = \sum_{\alpha,\beta=0}^{\infty} \sum_{\zeta=0,1} C_{\alpha\beta\zeta} T_{\alpha\beta\zeta}(\vec{R}) + 1. \tag{12}$$

Inserting (12) into the boundary condition (3) we get

$$\sum_{\alpha,\beta=0}^{\infty} \sum_{\zeta=0,1} C_{\alpha\beta\zeta} J_{\alpha\beta\zeta}(\hat{R}) = -\hat{R} \cdot (\vec{F} + 2\gamma Y \hat{e}_x) \Big|_{R=1}, \tag{13}$$

with $J(\hat{R})$ defined as

$$J(\hat{R}) \equiv \hat{R} \cdot \left( -\frac{1}{2} \frac{\partial}{\partial \vec{R}} + \vec{F} + 2\gamma \hat{e}_x Y \right) T(\vec{R}) \Big|_{R=1}. \tag{14}$$

## IV. LINEARIZATION IN $\vec{F}$

For small dimensionless external force $F^i \ll 1$, the boundary problem (13) and (14) can be linearized in $\vec{F}$. Each quantity $Q$ [$Q$ stands for $G_2(\hat{R}), T(\hat{R}), J(\hat{R}), C, \ldots$] is approximated by a linear function of $\vec{F}$,

$$Q = Q^{(0)} + \vec{Q}^{(1)} \cdot \vec{F}. \tag{15}$$

Equations (5)–(7) and (15) determine $\overleftrightarrow{M}(\gamma)$, the two-particle contribution to the self-mobility tensor, as a functional of $\vec{G}^{(1)}(\hat{R})$:

$$\overleftrightarrow{M} = \frac{3}{4\pi} \int_{S^2} d^2\Omega \hat{R} \vec{G}^{(1)}(\hat{R}). \tag{16}$$

($\overleftrightarrow{M}$ does not depend on $G^{(0)}$ due to symmetry.)

To find $\vec{G}^{(1)}(\hat{R})$ we are going to solve linearized Eqs. (13) and (14) for $C^{(i)}$, $i = 0, 1$. To this goal we apply the idea from [19]; namely, we expand each quantity $Q^{(i)}(\hat{R})$ ($Q$ stands for $T$ or $J$) into Cartesian harmonics $Y_{abc}(\hat{R})$ [24], with $a, b = 0, 1, 2, \ldots$ and $c = 0, 1$ (these harmonics form a complete set of functions on $S^2$):

$$Q^{(i)}(\hat{R}) = \sum_{a,b=0}^{\infty} Q^{(i)abc} Y_{abc}(\hat{R}), \tag{17}$$

$$Y_{abc}(\hat{R}) = -\frac{(-R)^{a+b+c}}{[2(a+b+c)-1]!!}\frac{\partial^a}{\partial X^a}\frac{\partial^b}{\partial Y^b}\frac{\partial^c}{\partial Z^c}\frac{1}{R}. \tag{18}$$

Each of the equations following from the linearization of the boundary problem (13) and (14), and therefore all the quantities $Q^{(i)}(\hat{R})$, are either symmetric or antisymmetric under the transformations $Z \to -Z$ and $(X,Y) \to (-X,-Y)$. In particular, it follows from symmetry in $Z$ that there is no summation over $c$ in Eq. (17): $c = 0$ or 1 for $Q^{(i)}$ symmetric or antisymmetric with respect to $Z \to -Z$. In addition, $Q^{(i)abc} = 0$ for $a+b$ even or odd, depending on symmetry of $Q^{(i)}$ with respect to $(X,Y) \to (-X,-Y)$.

After linearization the boundary condition (13) and (14) takes the form of an infinite set of algebraic equations for the coefficients $C^{(0)}$ and $\vec{C}^{(1)}$:

$$\sum_{\alpha,\beta=0}^{\infty} C^{(0)}_{\alpha\beta 0} J^{(0)ab0}_{\alpha\beta 0} = -2\gamma\delta_{a1}\delta_{b1}, \tag{19}$$

$$\sum_{\alpha,\beta=0}^{\infty} \vec{C}^{(1)}_{\alpha\beta c} J^{(0)abc}_{\alpha\beta c} = -\sum_{\alpha,\beta=0}^{\infty} C^{(0)}_{\alpha\beta 0}\vec{J}^{(1)abc}_{\alpha\beta 0} - \hat{R}^{abc}, \tag{20}$$

where $\hat{R}^{abc} = (\delta_{a1}\delta_{b0}\delta_{c0},\ \delta_{a0}\delta_{b1}\delta_{c0},\ \delta_{a0}\delta_{b0}\delta_{c1})$. Because of symmetry $c = 1$ for the $Z$ component of Eq. (20) and $c = 0$ for the other components.

## V. EXPANSION IN $\gamma^{1/2}$

The self-mobility dependence on the shear rate $\gamma$ needs careful analysis since the perturbation in $\gamma$, applied to the stationary Smoluchowski equation, is singular and $T^{(i)}$ are not analytical in $\gamma$. Equations (10) and (11) determine the scaling $T^{(i)} \sim \gamma^{n/2} B^{(i)}[\gamma^{1/2}\vec{R}]$, suggesting that $T^{(i)}$ could be expanded in $\gamma^{1/2}$. Indeed, it has been proved in [19, 22] that $T^{(i)}$ are analytic in $\gamma^{1/2}$.

To expand in $\gamma^{1/2}$ those Cartesian-harmonic projections of $\vec{G}^{(1)}(\hat{R})$, which are the components of $\overleftrightarrow{M}$ [see Eq. (22)], we make the following steps. First, starting from integral (10), we construct an algorithm to evaluate explicitly the expansion in $\gamma^{1/2}$ of the Cartesian-harmonic projections of $T^{(0)}$ and $\vec{T}^{(1)}$. Then, the expansion of $J^{(i)abc}_{\alpha\beta\zeta}$ follows immediately. Next, to calculate the expansion coefficients of $C^{(i)}_{\alpha\beta\zeta}$,

$$C^{(i)}_{\alpha\beta\zeta} = \sum_{\nu=0}^{\infty} C^{(i)}_{\alpha\beta\zeta}(\nu)\gamma^{\nu/2}, \tag{21}$$

we expand in $\gamma^{1/2}$ Eqs. (19) and (20). To begin, we solve Eq. (19) in the increasing order of $u$. For each $u$, it turns out that only a finite number of the coefficients $C^{(0)}_{\alpha\beta\zeta}(u)$ is nonzero; all nonvanishing coefficients are expressed as linear combinations of $C^{(0)}_{\kappa\rho\eta}(\nu)$ with $\nu < u$, determined in the previous orders. We substitute the evaluated $C^{(0)}$ into Eq. (20) and analogously solve the hierarchy for $\vec{C}^{(1)}(u)$. For details see [19, 20, 22]. The expansion of the coefficients $C$ and of the fundamental solutions $T$ in $\gamma^{1/2}$ is finally inserted into Eq. (12) to get the Cartesian-harmonic projections of the two-particle distribution at the contact $\vec{G}^{(1)}(\hat{R})$ as a power series in $\gamma^{1/2}$ [22].

## VI. SHEAR-RATE EXPANSION OF SELF-MOBILITY

The expansion in $\gamma^{1/2}$ of $\overleftrightarrow{M}(\gamma)$ immediately follows from Eq. (16) and the expansion in $\gamma^{1/2}$ of $\vec{G}^{(1)}(\hat{R})$, projected onto the corresponding Cartesian harmonics [22]

$$\overleftrightarrow{M}(\gamma) = \begin{pmatrix} G_x^{(1)100} & G_y^{(1)100} & 0 \\ G_x^{(1)010} & G_y^{(1)010} & 0 \\ 0 & 0 & G_z^{(1)001} \end{pmatrix}. \tag{22}$$

The leading behavior of $\overleftrightarrow{M}(\gamma)$ is

$$\overleftrightarrow{M}(\gamma) = \begin{pmatrix} -1 & -\frac{8}{15}\gamma & 0 \\ \frac{22}{15}\gamma & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix} + O(\gamma^{3/2}). \tag{23}$$

In Ref. [18] the same leading behavior has been obtained by linear response method. Szamel *et al.* evaluated the coefficients of the linear $\gamma$ correction analytically for hard spheres, in agreement with our present results, and numerically for particles interacting via the screened Coulomb potential (in this case the coefficients depend on the potential hardness). [However, there is a sign error in the normalization of the results given in Ref. [18]: the numerical results presented in Fig. 1 of [18] and the analytical results for hard spheres correspond to $\overleftrightarrow{\mu}/|\mu^{(0)}|$ and $\overleftrightarrow{D}/|D^{(0)}|$ rather than $\overleftrightarrow{\mu}/\mu^{(0)}$ and $\overleftrightarrow{D}/D^{(0)}$.]

The expansion of $\overleftrightarrow{M}$ in $\gamma^{1/2}$ up to the 47th order has been evaluated numerically [22]. The series converges for small $\gamma$, but it is divergent for $\gamma^{1/2}$ above about 0.8. To evaluate the self-mobility tensor for higher $\gamma$ we have calculated a sequence of diagonal $[N/N]$ Padé approximants with $N \leq 23$. The estimated accuracy of the results is within 1% for $\gamma$ up to 5 and within 0.1% for $\gamma < 2$.

The dependence of $\overleftrightarrow{M}$ on $\gamma$ is presented in Fig. 1. Note that the approximation (23) is valid for $\gamma < 0.01$, a range hardly visible on the scale of the picture. Even for relatively small $\gamma$ a significant number of terms in the expansion has to be included.

It is interesting to analyze $M_{xy}$ as a function of $\gamma$. $M_{xy}$ is positive for large $\gamma$, as one could expect from a simple analysis of a diffusionless suspension [22]. However, $M_{xy}$ changes sign at $\gamma \approx 0.4$ and becomes negative for smaller values of $\gamma$. This means that movement in the $x$ direction driven by the $y$ component of the external force has opposite orientation for small and large $\gamma$.

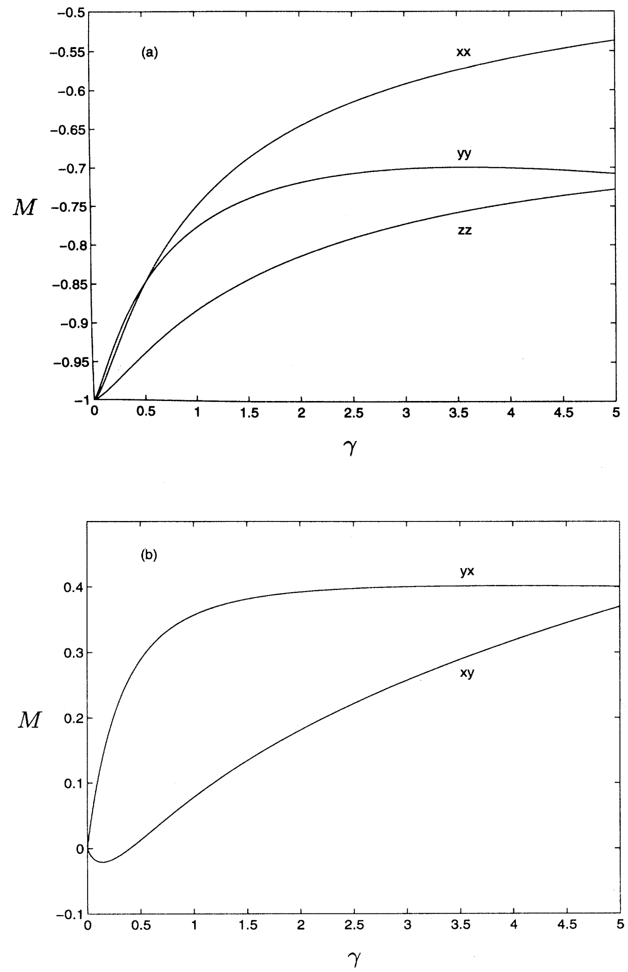


FIG. 1. Dependence of the self-mobility on the shear rate. (a) Diagonal and (b) off-diagonal components of $\overleftrightarrow{M}(\gamma)$, the two-particle contribution to the self-mobility tensor, are plotted as functions of $\gamma$. The curves are labeled with the corresponding component indices. Both $M$ and $\gamma$ are dimensionless.

## VII. CONCLUDING REMARKS

We have determined the shear-rate dependence of the self-mobility tensor $\overleftrightarrow{\mu}$ for a semidilute hard-sphere suspension with no hydrodynamic interactions. For shear rates $\gamma \neq 0$ the self-mobility tensor is nondiagonal and nonsymmetric.

As a result of the singular character of the perturbing convective term in the two-particle Smoluchowski equation the self-mobility tensor is not analytic in $\gamma$. However, the expansion in powers of $\gamma^{1/2}$ can be performed. The algorithm to determine the expansion coefficients has been constructed and used to determine numerically (by means of Padé approximants) the dependence of $\overleftrightarrow{\mu}$ on $\gamma$ in a range of $\gamma \leq 5$.

Similar techniques can also be applied to analyze the self-diffusion process. In this case one has to solve the Smoluchowski equation with no external force, but with the nonuniform density of untagged particles $n_s$. In the special case when $n_s$ does not change along the flow direction, the long-time long-wavelength limit can be performed in the standard way. Resulting formal expressions for the self-diffusion matrix coefficients $D_{ab}$ with $b \neq x$ ($\hat{x}$ is the flow direction), in terms of the solution to the two-particle Smoluchowski equation, have been derived in Ref. [18]. These expressions can be explicitly evaluated using the $\gamma^{1/2}$ expansion technique described in the present paper.

We note, however, that, in general (that is, when $n_s$ does change along the flow direction), the analysis of Ref. [18] is incorrect. In this case, even in the long-wavelength limit, the density $n_s$ changes, due to convection, on the time scale $\tau_s = \gamma_0^{-1}$, which for finite values of the shear Péclet number $\gamma$ [Eq. (9)] is comparable to the Boltzmann time scale $\tau_b$. The long-time analysis cannot be performed without taking this phenomenon into account. This was not noticed in [18] and the obtained results for $D_{ab}$ are incorrect if $b = x$. The long-time long-wavelength self-diffusion process for a colloidal suspension under shear is more complex and requires further analysis.

As the last remark we note that the shear-rate-dependent steady-state nonequilibrium structure and the transport properties have been studied not only for colloidal suspensions but also for simple fluids. For example, simple fluids under stationary shear has been recently investigated [25–29]. A review of some earlier results is given in [30].

For simple fluids, most of the work has been done theoretically and by means of computer simulations. Experimentally it is very difficult to achieve shear rates corresponding to intermediate or large Péclet number. This is due to the fact that the Péclet number appropriate for molecular systems is calculated using a molecular time scale in place of the Brownian time scale. Therefore, for a given shear rate, the Péclet number of a molecular fluid is much smaller than for a colloidal suspension. For simple fluids it is easier to make experimental observations in steady states generated by large temperature gradients. In particular, for such systems a nonlinear distortion of the structure factor from its equilibrium form has been measured in Rayleigh scattering experiments [31].

We have mentioned here molecular systems since there are important analogies between the behavior of colloidal suspensions and simple fluids undergoing strong shearing motion. In particular, the breakdown of the Einstein relation between self-diffusion and self-mobility tensors has been demonstrated for both systems (in [18] for colloids and in [27] for simple fluids).

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